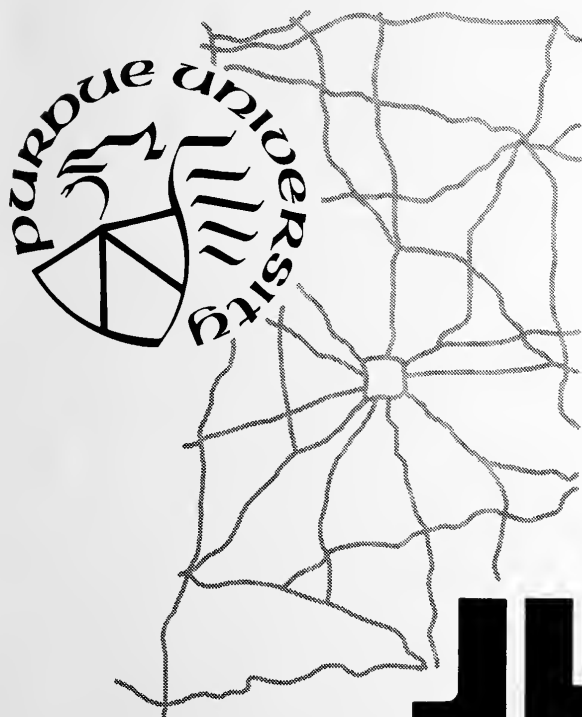


MECHANICAL BEHAVIOR AND REINFORCEMENT
OF MINERAL FILLER-ASPHALT MIXTURES

FEBRUARY 1973 - NUMBER 5



BY

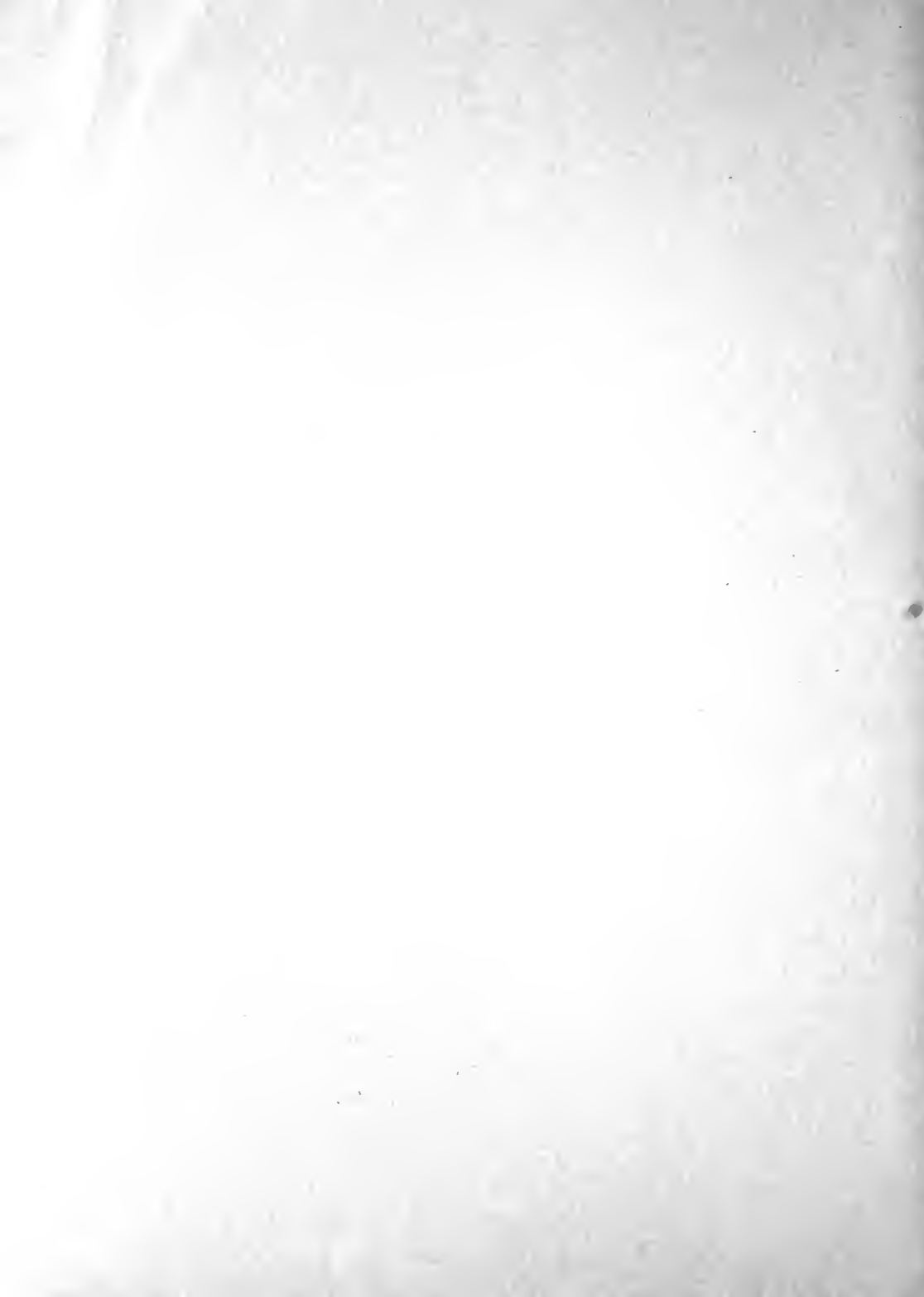
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INDIANA STATE HIGHWAY COMMISSION



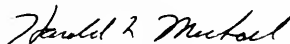
Technical Paper
MECHANICAL BEHAVIOR AND REINFORCEMENT OF
MINERAL FILLER-ASPHALT MIXTURES

TO: J. F. McLaughlin, Director February 8, 1973
Joint Highway Research Project
Project: C-36-6Y
FROM: H. L. Michael, Associate Director
Joint Highway Research Project File: 2-4-25

The attached Technical Paper titled "Mechanical Behavior and Reinforcement of Mineral Filler-Asphalt Mixtures" has been accepted for presentation at the Annual Meeting of the Association of Asphalt Paving Technologists in Houston, Texas, February 12-14, 1973. The Paper has been authored by David A. Anderson and W. H. Goetz and is from the Research Report titled "Mechanical Behavior of Asphalt-Mineral Powder Composites and Asphalt-Mineral Interaction" submitted as an Interim Report on the HPR Part II Research Study titled "Cracking in Bituminous Mixtures". That Report was accepted by all cooperating organizations.

The Paper is submitted to the Advisory Board for approval of publication. It will be submitted to the ISHC and FHWA for information and any review and comment they care to make. Inasmuch as the Interim Report from which the Paper came has been accepted by ISHC and FHWA, approval of presentation and publication is not required.

Respectfully submitted,



Harold L. Michael
Associate Director

HLM:ms

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Technical Paper

MECHANICAL BEHAVIOR AND REINFORCEMENT OF
MINERAL FILLER-ASPHALT MIXTURES

by

David A. Anderson and W. H. Goetz*

Joint Highway Research Project

Project No.: C-36-6Y

File No.: 2-4-25

Prepared as Part of an Investigation

Conducted by

Joint Highway Research Project
Engineering Experiment Station
Purdue University

in cooperation with the

Indiana State Highway Commission

and the

U.S. Department of Transportation
Federal Highway Administration

to be presented at

Annual Meeting Association of Asphalt Paving Technologists

Houston, Texas

February 12-14, 1973

Purdue University
West Lafayette, Indiana
February 8, 1973

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INTRODUCTION

Asphaltic concretes are mixtures of three components: mineral aggregate, asphalt, and air. The proportion of each component and the gradation of the aggregate is generally determined by one of several mixture design methods.⁽¹⁾ Additional constraints are generally placed on the individual components in terms of asphalt consistency and aggregate characteristics.

In the usual dense-graded asphaltic concrete the aggregate portion is well graded from the coarsest size down to the 200 mesh size (74 μm). The intent of the uniform gradation is to build an aggregate framework with maximum particle contact wherein each subsequent size just fills the voids of the larger size.⁽²⁾

Typically, the minus 200 material may be 5 per cent of the total mixture volume and the asphalt 13 per cent. Allowing some of the asphalt to be adsorbed or otherwise trapped in the plus 200 fraction, a minus 200-asphalt ratio as high as 40-60, by volume, is very realistic. At a 40-60 ratio the mineral is floating in the asphalt and particle-particle contact between the aggregate particles no longer predominates. The 40 value represents a solids concentration less than that at densest packing.^(3,4)

Under these conditions, the mixture consistency is controlled by the volume filling and physico-chemical reinforcing nature of the filler rather than by mineral to mineral contact. This is in contrast to the behavior of the overall mixture where a significant contribution to the

load-deformation behavior is made by the nature of the binder as well as by the nature of the particle-particle contact.^(5,6)

It is customary to specify a range of consistency for the asphalt used in asphaltic concrete. To assume that this in turn specifies a corresponding range of consistency in the filler-asphalt matrix is to imply that the consistency of the asphalt in the matrix is uninfluenced by the nature of the filler. In other words, the tacit assumption in customary usage is that there is an absence of interaction between the asphalt and the filler.

Review of Literature

Different filler-asphalt systems have been reported on in the literature, both as separate systems and as incorporated into asphaltic concrete. Perhaps one of the earliest references is to the work of Clifford Richardson.⁽⁷⁾ He postulated that the function of the filler was more than mere void filling, implying that some sort of physico-chemical phenomenon was operative. Richardson reported that silica, limestone dust, and portland cement made successively better fillers as they adsorbed a correspondingly thicker film of asphalt. It is interesting to note that when supplied as commercial fillers, silica, limestone dust, and portland cement is a very likely order of increasing fineness.

By the late 1930's many studies on the asphalt-mineral filler system had been completed in an attempt to characterize fillers with respect to their potential for stiffening the system. Traxler⁽⁸⁾ reported on a series of extensive investigations in which the findings were, essentially, (a) that the relative viscosity (η_{rel} , defined as mixture viscosity

divided by viscosity of the bitumen at zero solids concentration) for varying filler concentrations, c_v , was independent of the nature and consistency of the suspending medium, (b) that the stiffening effect of the filler could not always be adequately predicted by the per cent voids in the dry compacted filler, and (c) that only by mixing the filler and asphalt and testing it could a reliable prediction of the ensuing stiffening be made. Traxler considered size and size distribution as the fundamental filler parameters in that they affect the void content and average void diameter of packed powders. More recent articles by Traxler are in essential agreement with the earlier findings.⁽⁹⁾

Mitchell and Lee⁽¹⁰⁾ also conducted research in an attempt to find a parameter that would adequately predict the ability of a mineral filler to reinforce the asphalt to which it is added. Their data were taken for relatively small solid concentrations and indicated that the bulk settled volume of filler in benzene is a good predictor of the reinforcement to be expected. The actual experiment consisted of allowing different fillers to settle in benzene and adding equal bulk settled volumes of different fillers to a constant volume of asphalt: it was found that for small filler-asphalt ratios equal settled volumes gave equivalent reinforcement (η_{rel}). It has been demonstrated by others that the settled volume in benzene is directly related to fineness.

A very extensive series of experiments on mineral fillers and mineral filler-asphalt composites has been reported by Rigden.⁽¹¹⁾ His viscosity data were collected from experiments using both a Couette viscometer and a uniaxial tension test (on the more viscous material). Surface

area, settled volume in benzene, and permeability measurements (of the dry packed powder) were made.

At the higher volume concentrations no valid correlation between any of the primary physical properties of the powders and η_{rel} was found, nor did the settled bulk volume in benzene provide a valid correlation. By measuring the average hydraulic radius of the voids in the dry packed powder, \bar{r} , an improved correlation was possible by plotting $\log \eta_{rel}$ vs $c_v / \sqrt{\bar{r}}$. However, there were still deviations of over an order of magnitude in η_{rel} in this correlation. Rigden calculated η_{rel} for the different systems at the arbitrary shear stress of 6×10^4 dynes/cm sq. One of his figures suggests strains of 30 per cent in the testing, hardly a small strain.

According to the data presented by Rigden, at the higher filler concentrations, both the temperature susceptibility and the degree of complex flow are functions of the volume concentration.

Winniford⁽¹²⁾ used the sliding plate microviscosimeter to study filler-asphalt systems. Several different mechanisms were postulated in order to explain the stiffening effect of the filler. In addition to the mechanism provided by volume filling, they were, (a) gelation of the asphalt by the surface with attendant non-newtonian flow and lowered temperature susceptibility, (b) formation of thick viscous coatings which increased the effective solids concentration and (c) surface shielding by adsorbed asphaltenes.

Steady state viscosity values were obtained on fillers of differing size but of the same mineral type. Roughly it was shown that reinforcement was more pronounced with the smaller sized material: two 10 μ m and

25 μm CaCO_3 fillers gave essentially the same η_{rel} but this was not true of the more finely divided fillers.

Winniford also compared a surface-treated filler to an untreated filler. By treating the silica surfaces with dichlorodimethylsilane the relative viscosity of the systems was increased. The surface treatment was interpreted as disallowing an adsorbed layer of "asphaltic material" which, when present, shields the particles, lowering the interparticle van der Waals forces. Calcium carbonate was considered to strongly adsorb asphaltenes and in the process significantly increase its effective volume concentration.

Warden, et al⁽¹³⁾ presented data on filler-asphalt systems in conjunction with field observations. These authors, motivated by field failures which they attributed to filler type, sought an easily measurable parameter that would predict the performance of the filler in the pavement. The tests performed on the fillers were empirical tests currently in vogue in asphalt technology. A re-examination of the early work by Traxler proved once again inconclusive in that no one single parameter was capable of predicting the reinforcement to be expected from a given mineral filler. The softening point of the filler systems was found to be quite critical to filler type.

Recently Tunnickliff has written a review paper on the subject of mineral fillers.⁽¹⁴⁾ A substantial portion of his paper is devoted to a definition of mineral filler; it is his desire to define the filler as that part of the fine aggregate that acts as though it is a "part of the binder." It is suggested that the influence of the surface may be extended into the asphalt through a surface energy gradient. A later

paper by Tunnicliff⁽¹⁵⁾ contains another review and again concludes that the filler should be defined as that material suspended in the asphalt. These two papers by Tunnicliff contain a comprehensive listing of references on mineral fillers.

Heukelon and Wijga⁽¹⁶⁾ have developed a method for expressing the relative viscosity of dispersions as a function of volume concentration and rate of shear. Any increase in relative viscosity over that of volume filling alone is attributed to the degree of peptization of the particles and/or particle shape. Their approach is to use an experimentally determined constant, m , to correct the actual volume concentration, c_v . The value of m is considered a function of both the asphalt and the filler and is determined experimentally for each filler-asphalt combination.

Much of the work reported in the literature for filler-asphalt systems has been done by comparing steady state viscosity measurements of the filled system to those of the original unfilled bitumen (η_{rel}). This steady state viscosity is usually determined by applying a load to the sample in question and waiting for the resulting displacement versus time plot to become linear. The stress divided by the slope of this plot is then given as the coefficient of viscosity. Because most bitumens at their service temperatures exhibit elastic effects that retard the viscous flow mechanism, it is necessary to pursue large strains before viscous flow is approached. In addition, because the approach to linearity is exponential, it is very difficult to be certain exactly when the steady state situation has been obtained.

Even if it were possible to easily define a steady state condition, the η_{rel} approach cannot account for elastic effects: additional parameters are needed to characterize short term behavior. Except for perhaps long-term creep, the elastic effects are important in determining the response of the material in time and temperature regions often encountered in the pavement system. The use of η_{rel} by itself appears to be of useful but limited value and a more comprehensive characterization of the mineral filler-asphalt system is needed.

Statement of Problem

Based on the foregoing remarks additional research on the behavior of the mineral filler-asphalt matrix or composite was considered desirable. Two problems were considered; the first to find an adequate method for characterizing the mechanical behavior (stress-strain response) of the mineral filler-asphalt composite. Second, based on the mechanical characterization, the potential for interaction between mineral filler and asphalt was evaluated.

MATERIALS AND SAMPLE PREPARATION

Previous work on filled bituminous systems has, for the most part been done with commercial fillers. Such materials have included crusher dust, silt, flyash and so forth. These materials are by nature ill defined, both in terms of mineralogical composition and particle size distribution. The series of mineral powders used in this study were carefully prepared and sized. Two different asphalts, supplied by the Federal Highway Administration, were mixed with the powders using a specially developed mixing procedure.

Mineral Powders

Six different minerals were selected for use and prepared to give a well-defined size distribution. Quartz and calcite were used in the bulk of the work because they were readily available and are generally considered to represent the range of materials between acidic and basic commonly used as fillers in bituminous mixtures.

An x-ray diffraction pattern was determined for each of the finely divided mineral powders. Each powder produced a pattern in keeping with its description and no unexplained peaks were found.

All the mineral powders, except the quartz, were prepared by first crushing the original crystal fragments to a fine sand size. This material was then ground in a ball mill until the majority of it passed the 400 mesh sieve. The quartz was obtained as a powder but required considerable grinding before the majority of it would pass the 400 mesh sieve.

After grinding, each powder was separated into several different fractions by allowing very dilute suspensions of the powders to sediment for various periods of time. At the end of each time period the supernatant was decanted and the process repeated until the supernatant was clean. This process produced a series of clean, one-sized powders as described in Table 1.

The nominal size determined from a sedimentation process must be used with caution because of the many assumptions made for this process.^(17,18) With this in mind several alternative methods for characterizing particle size were used.

Air permeability measurements were used to determine the size and surface area of each powder. The air permeability method is based on the relationship between particle size and the rate of flow of a gas or liquid through a packed bed of the powder.⁽¹⁹⁻²¹⁾ The experimental procedure used in this study⁽²²⁾ was adopted from Kamak.⁽²³⁾ Table 1 gives the porosity of the packed beds as well as the size and surface area values calculated from the permeability data.

As a check on the air permeability measurements, several nitrogen sorption measurements were made.⁽²⁴⁾ These data are also given in Table 1.

In order to compare the physical appearance of each of the powders and as a rough check on particle size, scanning electron photomicrographs were taken for each powder. Typical photographs are shown in Figures 1 and 2. Sizes calculated from these photomicrographs agreed quite well with the data of Table 1. The photomicrographs show the materials to be one sized, angular in shape and quite clean (i.e. lacking in finer contaminants).

Description of Asphalts

The two different asphalts used in this study were AC-20 grade material furnished by Mr. James M. Rice of the Federal Highway Administration. The two materials, designated B-3603 and B-3056, were from a group of study asphalts reported on at length in the literature.⁽²⁴⁻²⁶⁾

Viscosity data were obtained on the supplied material at 77 F and 275 F. These values are reported in Table 2 and agree quite well with values reported by other investigators.⁽²⁴⁾ At 77 F the B-3603 asphalt behaves as a newtonian fluid in the sliding plate microviscosimeter while the B-3056 asphalt is definitely non-newtonian.

Preparation and Description of Mineral Filler-Bitumen Mixtures

A special technique was developed for mixing the asphalt and mineral powder. Initially, the materials were mixed by hand in a beaker immersed in a heated oil bath. The resulting mixtures were not homogeneous. When the material was examined, after it had cooled, small pockets of uncoated powder were invariably found. This problem was solved by combining and mixing the two components in a closed system under vacuum.⁽²²⁾ A schematic diagram of the mixing apparatus is given in Figure 3. First the asphalt was placed in the 10ml beaker and the mineral placed in the hopper. The system was then evacuated (3-4mm Hg) after which the powder was slowly added (buzzer) to the asphalt. Mixing temperatures were 330 F as determined by thermocouple measurements in the asphalt during mixing. After mixing the beaker of material was removed from the mixing chamber and poured into disc shaped silicone rubber

molds. All the mixtures containing mineral powder were in the proportion 40 powder to 60 bitumen by volume.

It was anticipated that the vacuum process would have a measurable effect on the viscosity of the asphalt, especially in view of the fact that it was exposed not only to the heat and vacuum but to the stirring action as well. Viscosity data were obtained on asphalt sampled after ten and forty minutes in the mixing chamber. Within experimental error, no change in the 0.05 sec^{-1} viscosity value for the B-3603 asphalt was observed. The coefficient of viscosity for B-3056 asphalt increased to 10.7 and 12.5 megapoises (from the original 10.5) after ten and forty minutes. The lack of a sizeable viscosity increase during the vacuum treatment was rather surprising but was substantiated by a similar trend in the compliance data. Both the quasi-static and dynamic (sinusoidal) compliance remained essentially unchanged after forty minutes in the vacuum chamber.

CHARACTERIZATION OF MECHANICAL BEHAVIOR

The primary objective of any mechanical characterization is to establish an appropriate relationship between stress and strain. In this study predetermined shear stresses (both quasi-static and sinusoidal) were applied to the specimen at three different temperatures and the resulting strain levels observed.

Test Procedures

The sample to be tested was mounted as a pair of equivolume thin discs (approximately 1.5-in. in diameter by 0.04-in. thick) sandwiched between three steel plates (stacked face to face). A schematic of the fixture is given in Figure 4. The base plate was fixed to the lower platen of the test machine while the load was applied through the center plate to the load cell extension. The fixture was designed to minimize the effect of eccentric loading and mounting stresses and to eliminate fixture strain from the measurement of shear strain in the specimen.

The assembled base and sample plates, with the tightened side bars were mounted in the testing machine in a controlled temperature water bath. After mounting and zeroing, the plates were loosened and the testing performed.

Each sample was observed under a sinusoidally varying load (dynamic) and a step load (creep). At each temperature the sinusoidal tests

were performed first followed by the creep tests. The complex compliance, \bar{J}^* , and the creep function, $J(t)$, were determined from these data.

An effort was made to perform all the testing at sufficiently low strains such that linear behavior was preserved. No attempt was made to ascertain the onset of non-linear behavior because after each test point the samples were subsequently used for another data point. It was feared that exposure to the non-linear region might irreversibly affect subsequent behavior.

Occasionally, during the dynamic testing, a load sufficiently large to cause distortion in the Lissajous figure (x-y display of displacement-load) was applied. Although this did not occur with sufficient frequency to establish a definite pattern, it would appear that the onset of non-linearity is a function of both time and temperature. Linearity was observed over a wider range of strains and/or stresses at longer times and higher temperatures.

Dynamic Measurements

The dynamic data were obtained by applying a sinusoidally varying stress,

$$\sigma(\omega) = \sigma_0 \sin \omega t$$

to the test samples. $\sigma(\omega)$ is the stress in dynes per square centimeter, σ_0 the peak to peak stress, ω the angular frequency and, t , the time in seconds. If a material is linear viscoelastic, the response must also be sinusoidal but lagging the input by a phase angle, δ .⁽²⁷⁾

The response can then be written as

$$\gamma(\omega) = \gamma_0 \sin(\omega t - \delta)$$

where $\gamma(\omega)$ is strain in cm per cm, γ_0 the peak to peak strain, and δ the phase angle in radians.

It is convenient to write the dynamic property that relates stress to strain as a complex variable, \bar{J}^* , where, (28)

$$\bar{J}^*(\omega) = \frac{\gamma_0 e^{i(\omega t - \delta)}}{\sigma_0 e^{i\omega t}}$$

and

$$\bar{J}^* = \frac{\gamma_0}{\sigma_0} \cos \delta - \frac{\gamma_0}{\sigma_0} i \sin \delta \quad . \quad . \quad . \quad . \quad 1$$

Equation 1 separates the complex compliance into two parts, an in-phase component, J' and an out-of-phase component, J'' , defined as

$$J' = \frac{\gamma_0}{\sigma_0} \cos \delta \quad . \quad . \quad . \quad . \quad 2$$

$$J'' = \frac{\gamma_0}{\sigma_0} \sin \delta \quad . \quad . \quad . \quad . \quad 3$$

where,

$$|\bar{J}^*| = \frac{\gamma_0}{\sigma_0} \quad . \quad . \quad . \quad . \quad 4$$

The real part, J' , is commonly called the storage compliance because it represents stored or recoverable energy. The imaginary part, J'' , represents lost, nonrecoverable energy and is commonly called the loss compliance.

Equations 2, 3, and 4 were used to calculate the three components of compliance. The phase angle, δ , was calculated as the inverse tangent of J''/J' . The compliance data were additionally reduced to a reference temperature, T_0 , by multiplying by the ratio T/T_0 where T is the test temperature and T_0 is arbitrarily taken as 298 K. ⁽²⁷⁾ Theoretically the reduction should include a correction for density change ⁽²⁷⁾ but due to the limited temperature change this correction was omitted. Because the numerical values of compliance and time vary so widely, it is customary to consider the logarithm of the values. The time value is given as $1/\omega$; dimensionally $1/\omega$ is equivalent to time, and is roughly equal to time, t , in the creep experiment. A typical plot for each of the various compliance components and phase angle is given in Figures 5 through 8.

A necessary condition for linearity is the independence of $|\bar{J}^*|$ on the load level at any given frequency and temperature. ⁽³⁰⁾ This condition was shown to be valid by preparing plots of maximum stress versus maximum strain for those test points where data were taken at more than one stress level. The reciprocal of the slope of this curve defines $|\bar{J}^*|$ and, to the degree that the curve is linear, $|\bar{J}^*|$ is independent of stress level. A typical plot of this type is shown in Figure 9. From this plot, and others like it, a good indication of linearity was shown. Both J' and J'' were also found to satisfy this same linearity criterion.

As noted above⁽²⁷⁾ if a material is LVE a sinusoidally induced stress (input) must produce a sinusoidally varying strain (output). The sinusoidal data were recorded as x-y plots of displacement versus load. The figure produced from these plots is often called a Lissajous pattern (ellipse) and the shape of the pattern is rather sensitive to distortion in either the x or y input. In all cases the testing produced well defined ellipses, further substantiating the LVE character of the material.

In the dynamic testing, in all cases, steady state displacement was obtained after only a few cycles at the predetermined load, and repeated cycling did not change the level of the displacement. In a few instances prior levels of stress and frequency were reapplied after testing at other stress and frequency levels. In each case the original output was obtained within experimental error. This implies the absence of any thixotropy or structural breakdown in the material and, concomitantly, any artifacts in the data due to test history.

Creep Measurements

The creep compliance, $J(t)$, gives the relationship between shear strain and stress according to

$$J(t) = \frac{\gamma(t)}{\sigma_0} \quad . \quad . \quad . \quad . \quad 5$$

where the stress, σ_0 , takes the form of a step function applied at $t = 0$. Values of $J(t)$ were calculated according to Equation 5 and reduced to a reference temperature of 298 K. A typical plot of reduced unshifted creep data is given in Figure 10.

Two checks on the linear viscoelastic character of the creep data were imposed before the data were shifted. A necessary condition for LVE behavior is that the creep compliance be independent of stress.⁽²⁷⁾ This was found valid for creep data using the same technique described above for $|\bar{J}^*|$.

A second necessary condition involves an application of the Boltzmann superposition principle. Assuming the additivity of stress history as expressed by the superposition principle, the values of creep compliance were extended by adding the creep and recovery curves at equivalent times of creep and recovery respectively. The result of this construction is shown in Figure 10. The open circles represent the predicted data point whereas the solid points represent actual recorded data. To the extent that they form smooth curves, the predicted data points accurately predict the shape of the creep curve, supporting the condition of superposition.

Time-Temperature Superposition

A substantial simplification in the characterization of mechanical behavior may be obtained by applying the principle of reduced variables or time-temperature superposition.^(31,32) With an appropriate shifting of the curves along the time axis, the various curves, obtained at different temperatures, can be made to coincide, generating one smooth "master curve". The series of shift factors, defined as the various distances shifted, should yield a smooth continuous "shift function", $a_T(T-T_0)$, which is a function of temperature only. In this work, the technique was applied

to the values of $|\bar{J}^*|$ and smooth curves were generated without any vertical shift.

A plot of experimentally determined values of $\log a_T$ versus T for the two different asphalts used in this study is given in Figure 11. The values plotted are the averages for all the mixtures made with the two different asphaltic materials. There was no significant difference in the a_T function for the filled and unfilled asphalts. At the volume concentration used in this study, the shift function appears unaffected by the presence of the mineral.

While the matching of the curves of plotted data is a necessary criterion for the applicability of time-temperature superposition, two other criteria should be tested when possible.⁽²⁷⁾ The one a_T function should be unique to all the viscoelastic functions and the a_T function for the material in question must be consistent with experience. The uniqueness criterion was tested by applying the a_T values generated from the $|\bar{J}^*|$ data to J' , J'' , $\tan \delta$, and the creep function, $J(t)$.

Results are shown in Figures 12 and 13 for typical dynamic and creep data, respectively, where a single a_T function is shown to successfully shift both J' and J'' and the creep compliance.

A widely accepted functional form of a_T is given by the WLF equation as⁽²⁷⁾

$$\log a_T = \frac{-8.86 (T - T_g)}{101.6 + T - T_g} \quad . \quad . \quad . \quad . \quad 6$$

where the reference temperature, T_g , is an adjustable parameter, generally about 50 C above the glass transition temperature, T_g . The empirical

constants 8.86 and 101.6 represent data from many different polymer systems. Equation 6 implies a unique a_T function for all materials, provided they are referenced to the appropriate reference temperature, T_s .

Figure 14 shows the results of shifting (horizontally and vertically without rotation) the values from Figure 11 so that they best fit on the empirical equation for a_T as predicted by Equation 6.

Experimentally, for many different materials, including asphalts, it has been found that T_s is approximately 50 C above the experimentally measured glass transition temperature, T_g . (27, 31, 33-35) Values of T_s were approximately 36 C and 42 C respectively for the B3603 and B3056 asphalts and for the mixtures made with them. From the data presented, T_s is not influenced by the presence of the mineral filler but, as expected, does vary with the different asphalts.

COMPARISON AND DISCUSSION OF RESULTS

In the previous section it was shown that asphalt - mineral filler mixtures may be characterized as linear viscoelastic materials. In the following section a linear viscoelastic characterization will be used to evaluate the influence of mineral filler on the mechanical behavior of filler - asphalt mixtures.

Comparison of Quazi-static, Sinusoidal and Viscosity Results

A number of different methods are available for relating creep and dynamic data. (27,29,36-38) By manipulating the different viscoelastic variables, exact functions that relate the creep and dynamic compliance components can be written, either in terms of each other or in terms of the retardation function. In the latter case⁽²⁷⁾

$$J' = J_g + \int_{-\infty}^{\infty} \frac{L(\tau)}{1+\omega^2\tau^2} d\ln \tau \quad . \quad . \quad . \quad . \quad . \quad 7a$$

$$J'' = \int_{-\infty}^{\infty} \frac{L(\tau)\omega\tau}{1+\omega^2\tau^2} d\ln \tau + \frac{1}{\omega\eta} \quad . \quad . \quad . \quad . \quad . \quad 7b$$

These equations pose several problems: $L(\tau)$ must be defined and integrated over its entire range and values must be available for both J_g and η .

In order to obtain relations that are suitable for numerical calculation, certain simplifications and/or assumptions must be made in equations like 7, or alternative means for expressing the moduli must be sought. In the latter approach, for example, a series of exponentials may be used to advantage.⁽³⁷⁾

Most simply, van der Poel^(39, 40) has shown that good equivalence is obtained by setting

$$|\bar{J}^*|_{t=1/\omega} \approx J(t)$$

Although the results are not shown here, this relationship gave good predictions of the creep data with the largest error, approximately twenty percent, occurring in the transition region.

Ninomiya and Ferry⁽⁴¹⁾ have, by operating on the retardation function, derived the approximate relation where $\omega = \frac{1}{t}$:

$$J(t) = J'(\omega) + 0.40 J''(0.40 \omega) - 0.14 J''(10 \omega) \quad . \quad . \quad . \quad 8$$

This equation was used in this study to predict the creep compliance from the loss and storage compliance. A typical result is shown in Figure 13 where the solid line represents the results of equation 8. Over the available range of data, the calculated points were in excellent agreement with the recorded creep data.

Steady State Viscosity and Equilibrium Compliance

Unfortunately, the presence of significant retarded elasticity prohibits a straightforward value of steady state viscosity from the creep test unless it is allowed to proceed for a relatively long period of time. Referring to Equation 9,

$$J(t) = J_g + \int_{-\infty}^{\infty} L(\tau)(1-e^{-t/\tau}) d \ln \tau + t/\eta \quad . \quad . \quad . \quad 9$$

the plot of log compliance versus log time will attain a unit slope only when the sum of J_g and the integral (J_e) are small relative to t/η .

Ninomiya⁽⁴²⁾ has suggested a convenient method for calculating approximate values for both the steady state viscosity, η , and the steady state compliance. The results of these calculations are given in Table 3. The estimated value of η for the unfilled B3603 asphalt is listed as 6 megapoises, compared to 3.9 for the sliding plate microviscosimeter. The estimated value for the unfilled B3056 asphalt is 32 megapoises as compared to 10.5 measured in the microviscosimeter. To the extent that these values are valid they reflect the severe degree of non-newtonian flow evident in the B3056 asphalt.

The data for the B3056 mixtures made with SiO_2 show a greater decrease in compliance and increase in viscosity than those made with the CaCO_3 . (Recall that a decrease in J implies a "stiffening".) No values of J_e are given for the plain B3603 asphalt and for the B3603 - CaCO_3 mixtures, indicating that the t/η effect (flow) was large in comparison to the effect of J_e .

The viscosity of the B3056 mixtures is apparently particle size dependent, increasing with decreasing size. The ratio of filled to unfilled coefficient of viscosity, η/η^0 , ranges to, and above, 200 for the B3056 mixtures (see Table 3) but is only about 7, and independent of size, for the B3603 mixtures. This would indicate that the asphalt in the B3603 mixtures is only slightly reinforced* (4 compared to 7) but that the asphalt in the B3056 mixtures is strongly reinforced (4 compared to 200).

Comparison of Different Mineral Filler - Asphalt Mixtures

For the purposes of comparison, the creep compliances from each of the B3056 and B3603 asphalts are given in Figures 15 and 16 respectively. The general shape of the unfilled curve is retained upon the addition of the mineral filler. However, a simple vertical shift was found inadequate to bring the filled and unfilled moduli into coincidence. The B3603 mixtures can be brought into coincidence with both a vertical and horizontal shift but even this is inadequate for the B3056 mixtures.

The merging nature of the curves is striking at the small values of time. Figures 16 and 17 suggest that the degree of reinforcement for the glassy compliance, J_g , is unique for all mixtures, regardless of filler size and type or of asphalt type. This point is further emphasized in Figure 17, where relative values of compliance and coefficient of viscosity are given. $J^0(t)/J(t)$ is merely the ratio of the

*Borrowing terminology from work with filled elastomers,⁽⁴³⁾ a filler is said to be reinforcing if the composite formed with the filler has some enhanced physical property greater than that due to only the volume filling effect of the filler.

unfilled to filled compliance and η/η^0 is the ratio of the steady-state filled to unfilled viscosity (Table 3). The B3603 mixtures all fall on a single curve while the B3056 mixtures diverge at the longer test times.

Considerable attention has been given to the problem of predicting the moduli of time dependent particulate composites from the volume fraction of their components.⁽⁴⁴⁻⁵¹⁾ Above a few percent solids, the problem becomes extremely complex and empirical relationships prevail. From the various empirical equations and results given in the literature a stiffening effect of about four times should be expected for a non-reinforcing filler at the volume concentration as used in this study (0.40). Furthermore, it can be shown that the ratio of stiffening offered by the steady-state viscosity should also be realized in the moduli, independent of time and temperature.^(27, 51) Referring to Figure 16, this degree of stiffening is observed for the B3603 asphalt but the B3056 mixtures are definitely stiffened more than that expected from the volume filling effects alone.

The degree of reinforcement exhibited by the B3056 asphalt is dependent on time, size and nature of the mineral filler. Even the most interacting mixture (0.63-1.25 quartz) exhibits little or no reinforcement at the short loading times as $J(t)$ approaches J_g , suggesting that the reinforcement mechanism is a flow associated phenomena. This is further evidenced by the J_e data of Table 3, where the J_e is relatively uninfluenced by variations in mineral type.

The data shown in Figure 17 suggest that quartz is more reinforcing than calcite, the effect being more pronounced the smaller the size of

the filler. No hypothesis is offered to explain the reinforcement evidenced in the different mixtures except to suggest the presence of some sort of physico-chemical interaction. This statement is supported by the time, temperature and size (surface area) dependency of the observed reinforcement. This reinforcement must extend at depth into the asphalt. An increased particle size due only to adsorption at the surface would appear to be an inadequate explanation. The size of the rigidly adsorbed layer necessary to produce the 200 fold stiffening would be unreasonably thick.

CONCLUSIONS

Based on the data presented it can be concluded that both filled and unfilled asphalts can be satisfactorily represented as thermorheologically simple linear viscoelastic materials. Using this representation as a means for comparison it was shown that there is definitely a potential for reinforcement in mineral filler-bitumen composites. The extent of the reinforcement is dependent on the time of test, nature of the asphalt and type and size of the mineral. The reinforcement disappears at short loading times; the reinforcement mechanism appears to be a flow dependent phenomena. Due to the potential for reinforcement, that is interaction beyond volume filling along, it would appear that the consistency or stress-strain response of bulk asphalt is a poor descriptor or the asphalt-mineral filler matrix present in most asphaltic concrete.

TABLE 1
PROPERTIES OF SPECIALLY PREPARED FINELY DIVIDED MINERAL POWDERS

Material	Nominal Sedimentation Size, μm	Porosity	From Air Permeability		N_2 Sorption Surface Area, sq m/gram
			Size, μm	Surface area, sq m/gram	
Quartz	10 - 20	0.51	7.3	0.31	
	2.5 - 5.0	0.51	2.1	1.1	1.5
	0.63- 1.25	0.54	.59	3.8	4.4
Calcite	10 - 20	0.51	6.9	0.32	
	2.5 - 5.0	0.51	2.1	1.1	
	0.63- 1.25	0.47	1.4	1.6	2.4

TABLE 2
ASPHALT PROPERTIES

Sample	B 3056	B 3603
Viscosity, centistokes, 275 F	431	309
Viscosity, megapoises, 77F, 0.05 sec ⁻¹	10.5	3.9
Penetration, 77 F, 100g, 5 sec	34	45
Ductility, 77 F, 5cm/min	250+	250+

TABLE 3

CALCULATED COEFFICIENT OF VISCOSITY AND EQUILIBRIUM COMPLIANCE

Mineral			Viscosity ^a		Equilibrium Compliance ^a	
Asphalt	Mineral	Size, μm	η , 10^6 poises	Ratio: η/η^o	J_e , $10^{-8} \frac{\text{cm}^2}{\text{dyne}}$	Ratio: $\frac{J_e^o}{J_e}$
B3056	Unfilled	-----	32^b	1	80	1
	Calcite	10-20	530	17	8	10
		2.5-5.0	2000	62	6	13
		0.63-1.25	2500	78	3	26
	Quartz	10-20	1300	41	8	10
		2.5-5.0	3300	103	3	26
		0.63-1.25	6700	210	3	26
B3603	Unfilled	-----	6^c	--	--	--
	Calcite	10-20	44	7	--	--
		2.5-5.0	42	7	--	--
	Quartz	10-20	42	7	16	--
		2.5-5.0	47	8	9	--

a superscript, o, indicates unfilled asphalt

b B3056, Sliding plate microviscosimeter, 0.05 sec^{-1} , 10.5×10^6 poisesc B3603, Sliding plate microviscosimeter, 0.05 sec^{-1} , 3.9×10^6 poises

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Figure 1. Scanning Electron Photomicrograph,
 $10\text{--}20\ \mu\text{m}\ \text{SiO}_2$, $3000\times$

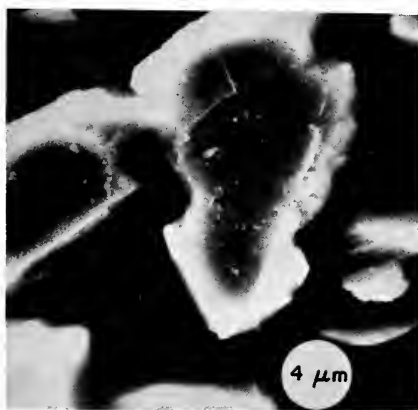


Figure 2. Scanning Electron Photomicrograph,
 $10\text{--}20\ \mu\text{m}\ \text{CaCO}_3$, $3000\times$

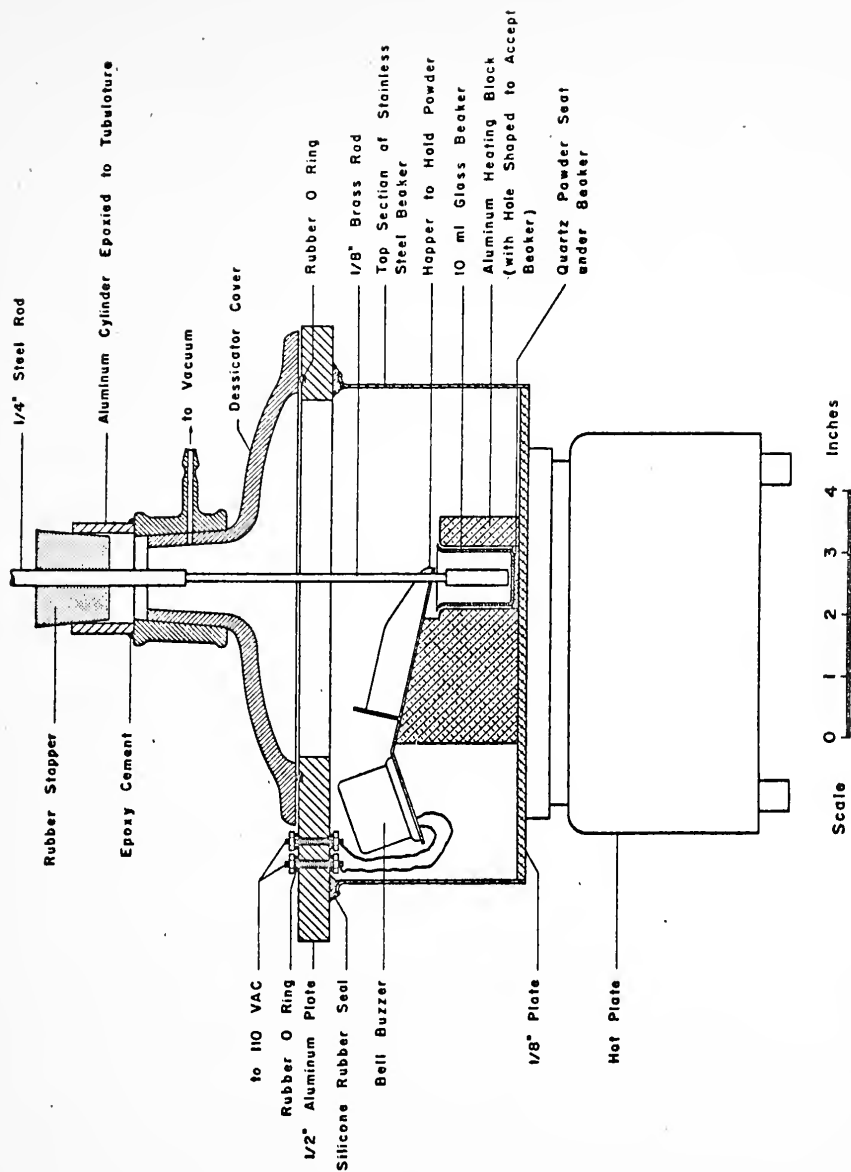


Figure 3. Schematic of Mixing Equipment.

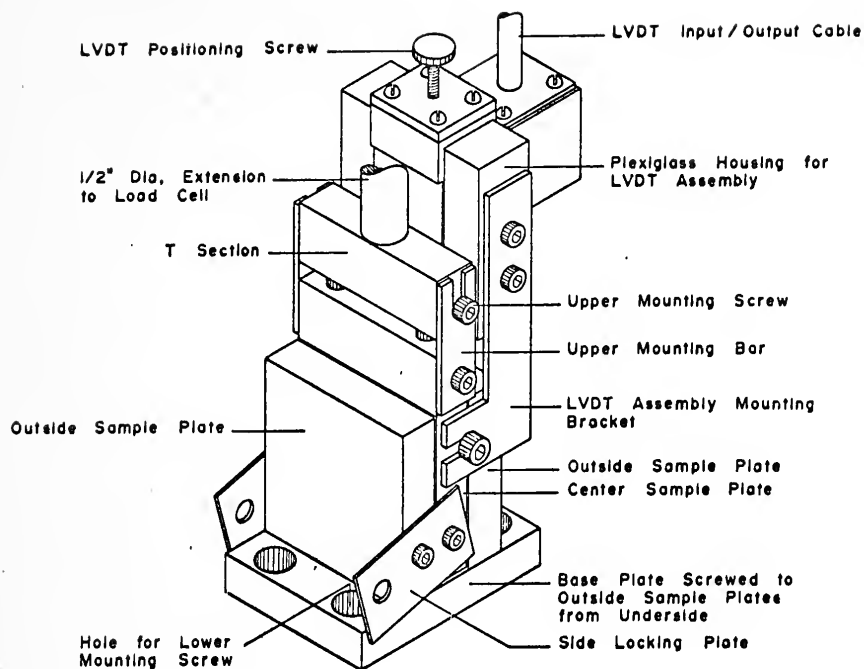


Figure 4. Assembled Sample Plates.

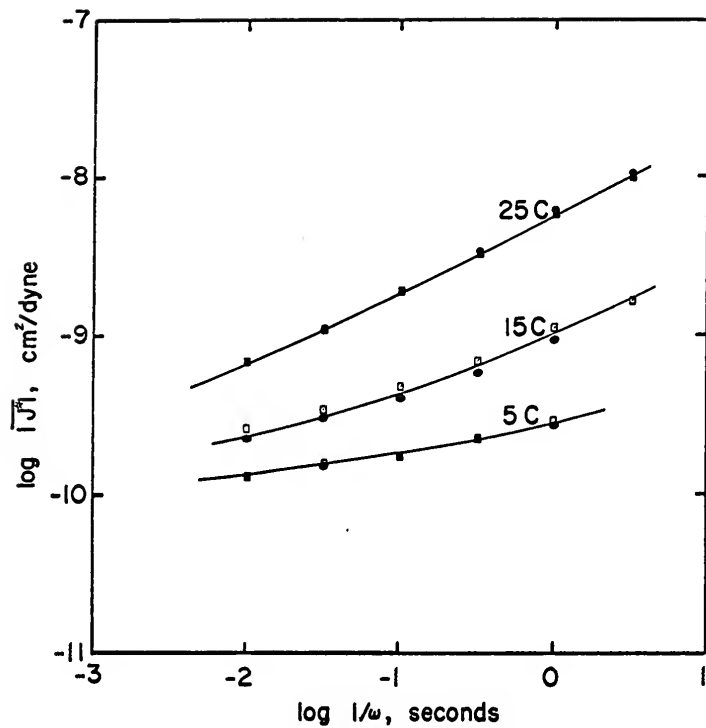


Figure 5. Typical Plot \bar{J}^* versus $\log 1/\omega$
 B-3056 Asphalt, 2.5-5.0 μm CaCO_3

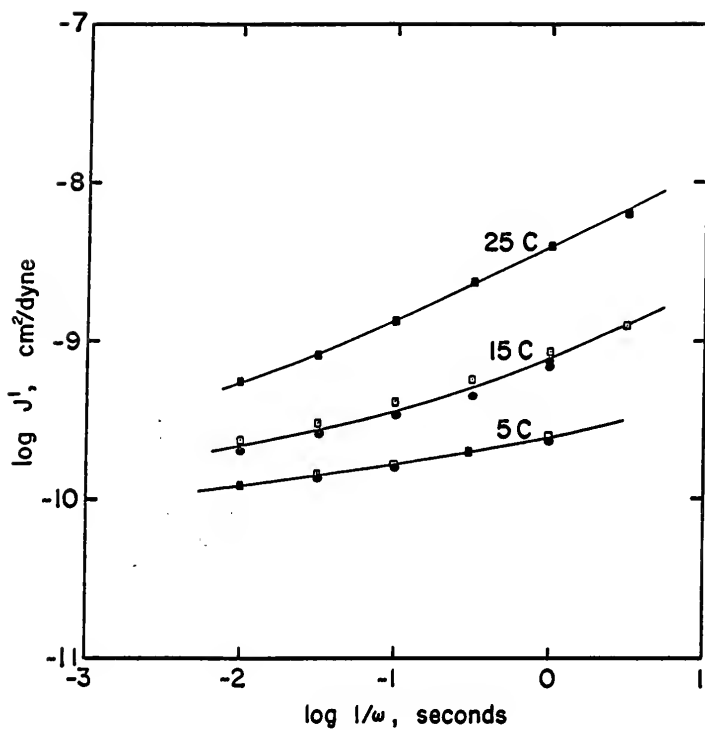


Figure 6. Typical Plot J' versus $\log 1/\omega$,
B-3056 Asphalt, 2.5-5.0 μm CaCO_3

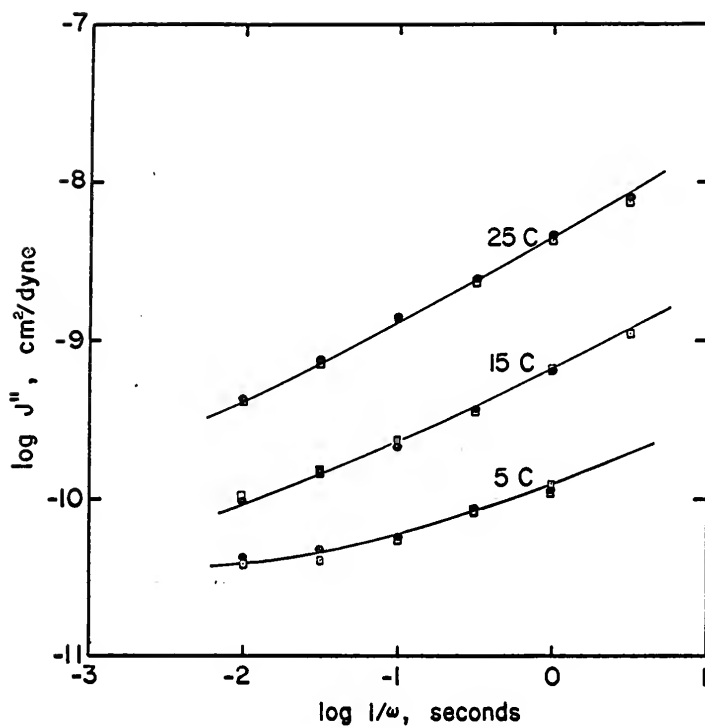


Figure 7. Typical Plot J'' versus $\log 1/\omega$,
B-3056 Asphalt, 2.5-5.0 μm CaCO_3

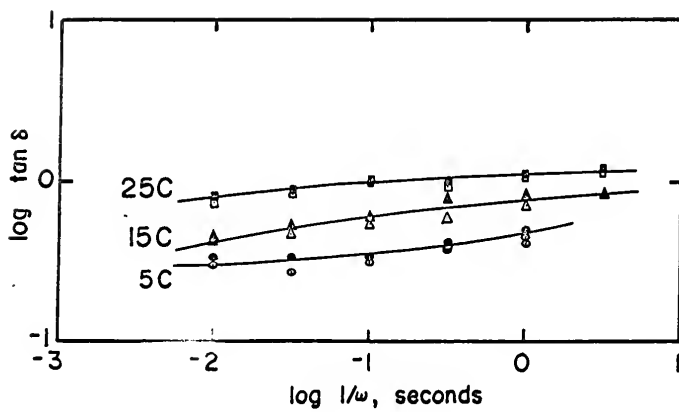


Figure 8. Typical Plot $\tan \delta$ versus $\log 1/\omega$
B-3056 Asphalt, 2.5-5.0 μm CaCO_3 .

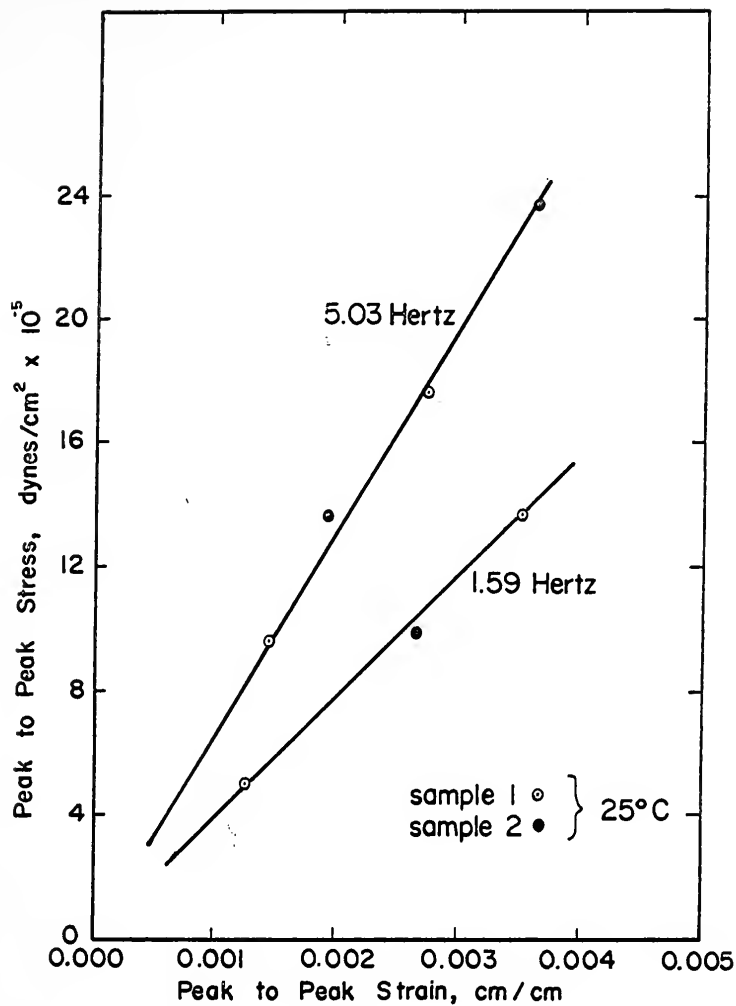


Figure 9. Peak to Peak Stress versus Strain,
B-3056 Asphalt, 10-20 μm CaCO_3

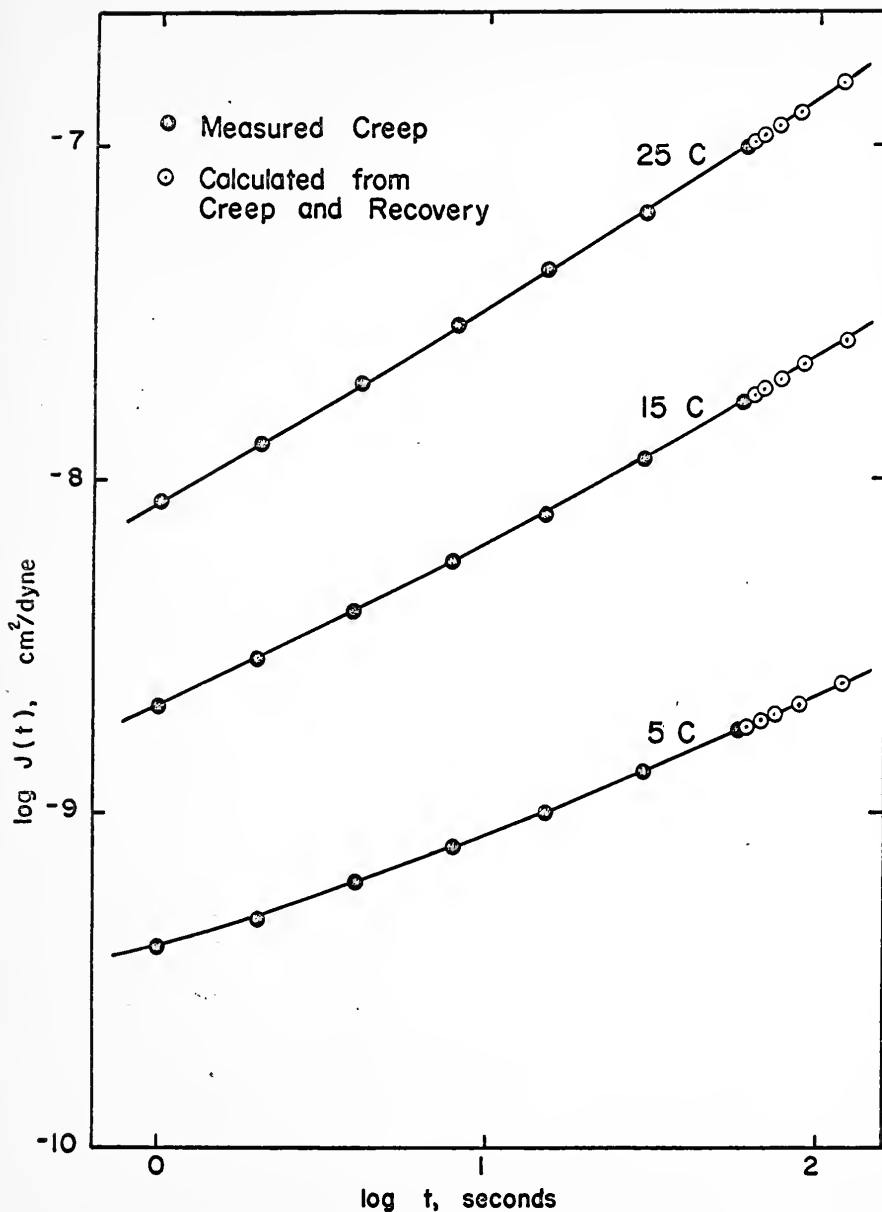


Figure 10. Recorded and Calculated Creep Compliance, B-3056 Asphalt, 2.5-5.0 $\mu\text{m S}_{12}\text{O}_2$

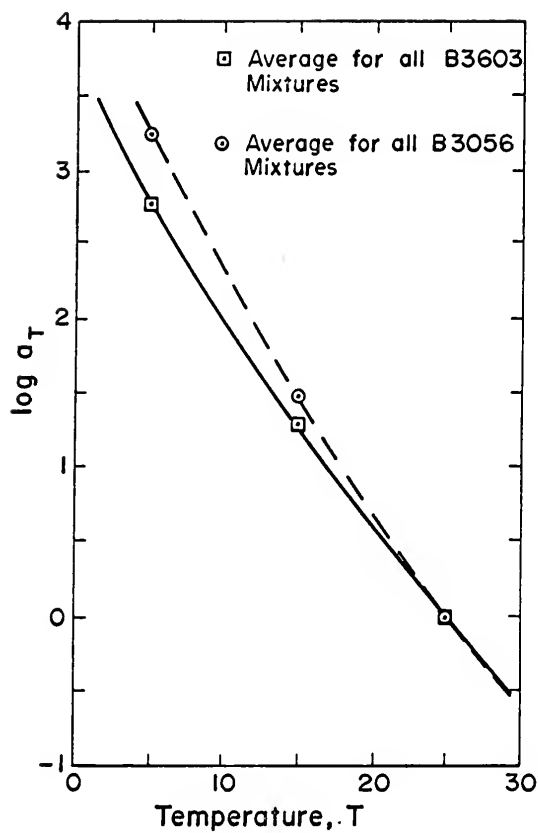


Figure 11. Experimentally Determined a_T .

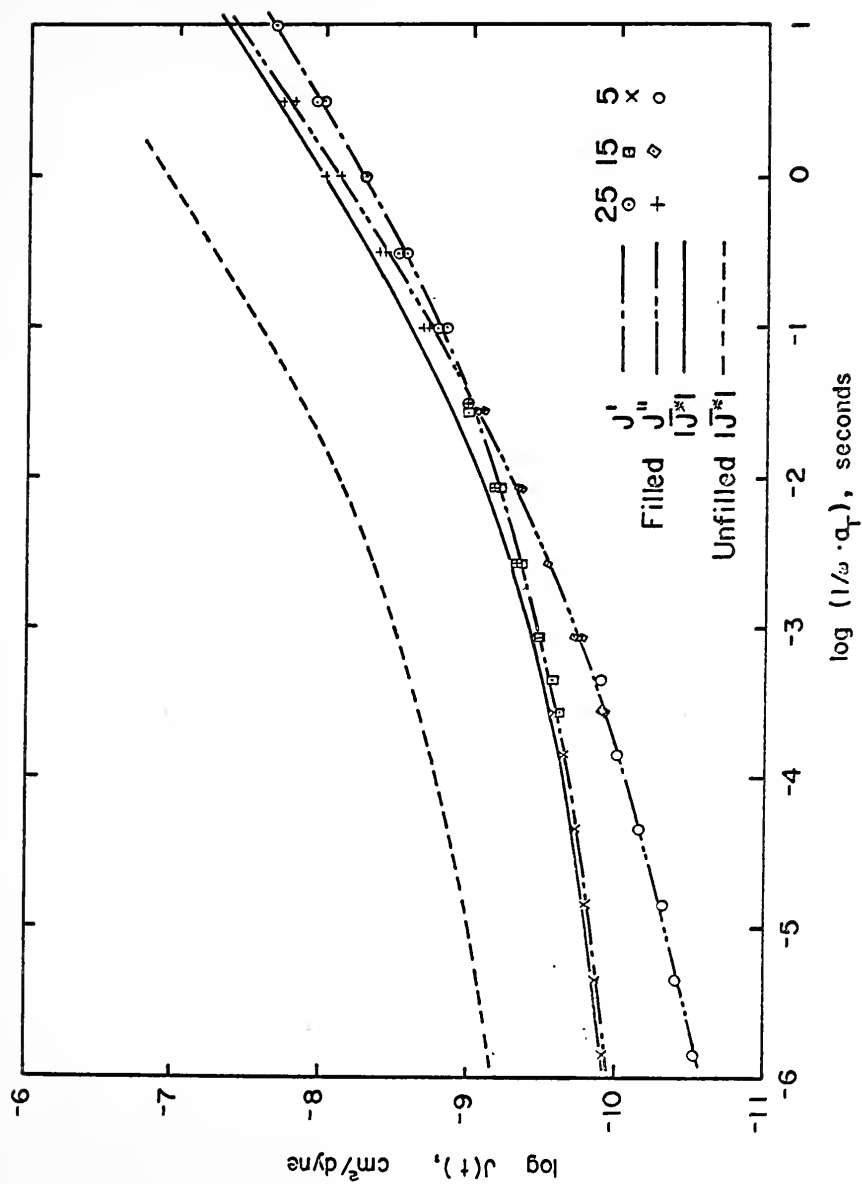


Figure 12. Dynamic Compliance Components, B-3056 Asphalt, 10-20 μm CaCO_3

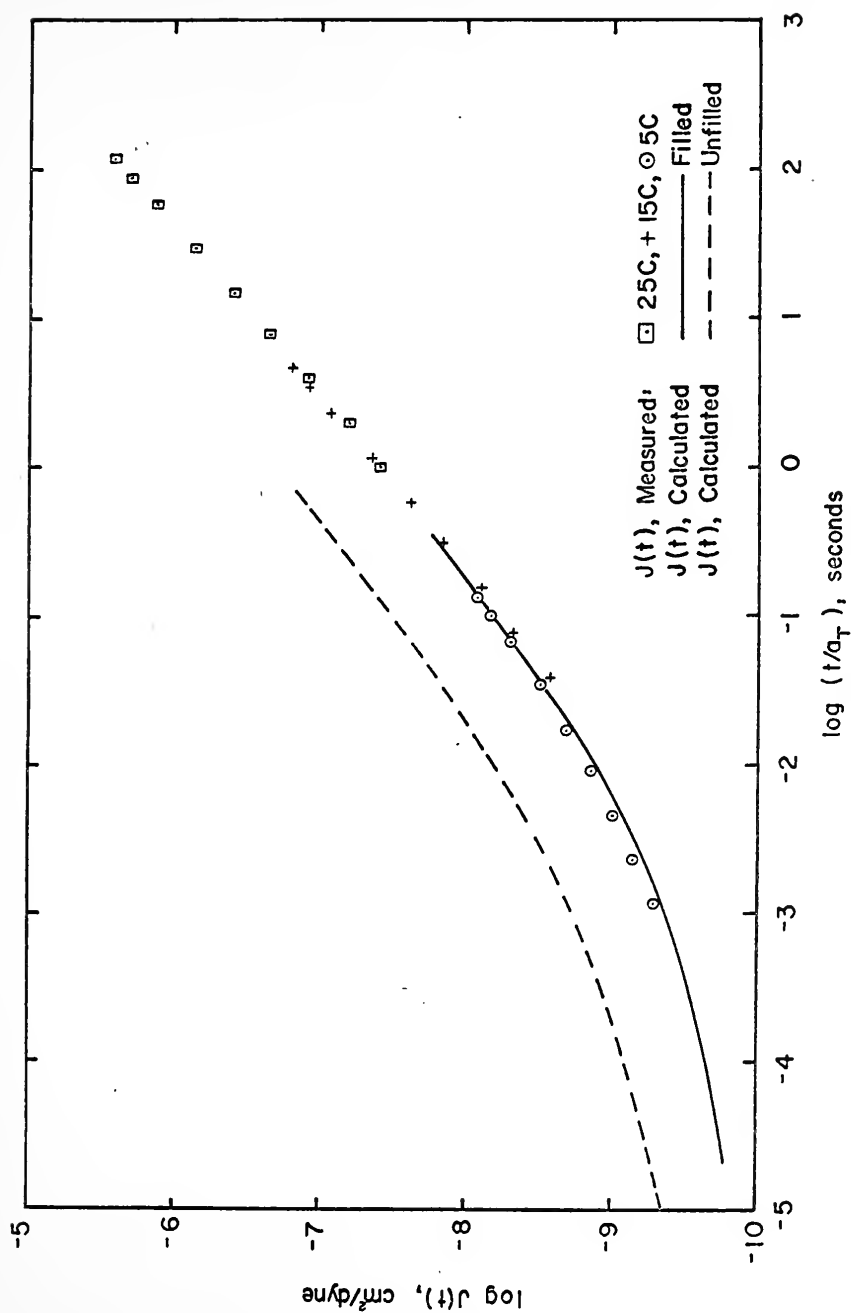


Figure 13. Creep Compliance, B-3603 Asphalt, 10-20 μm SiO_2

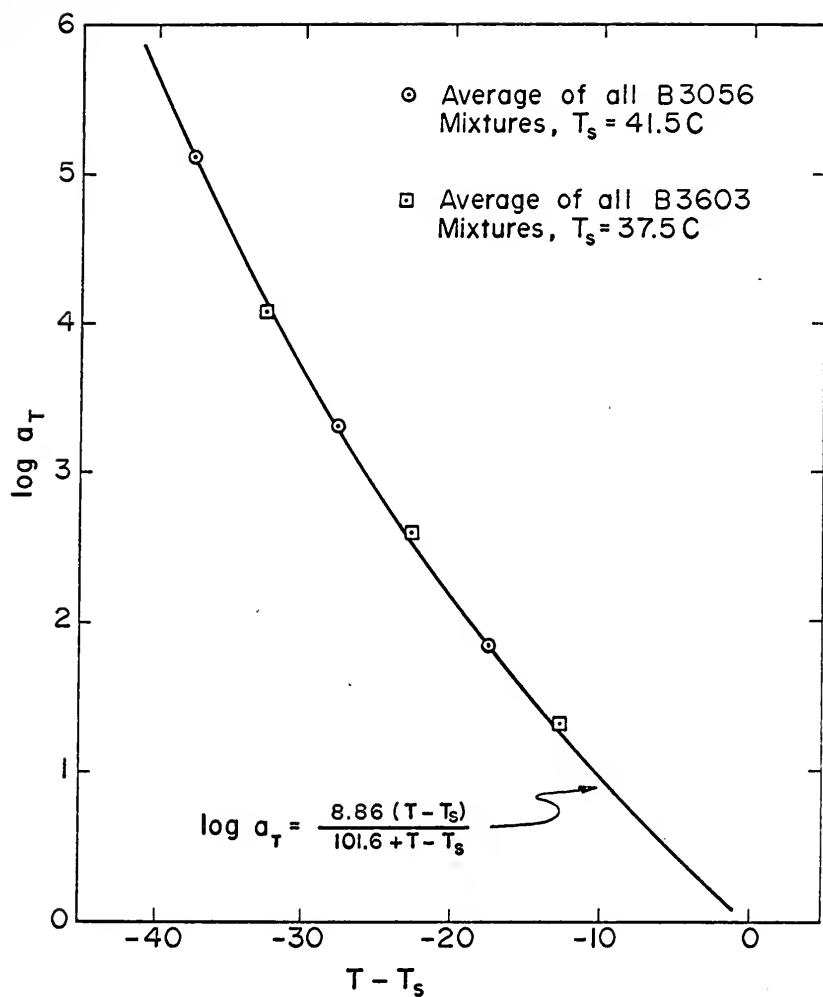


Figure 14. Shift Function, a_T , as a function of $T - T_s$

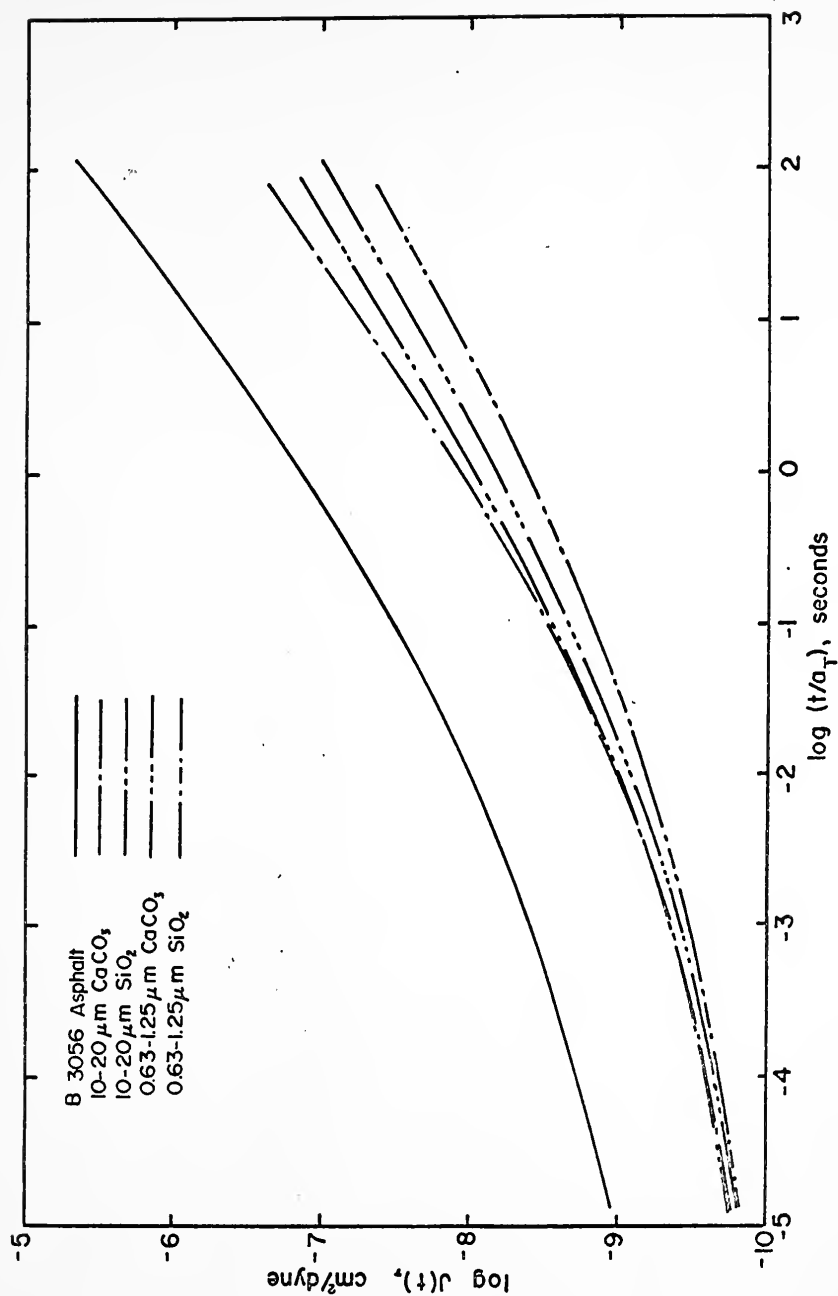


Figure 15. Creep Compliance, SiO_2 and CaCO_3 Mixtures, B-3056 Asphalt

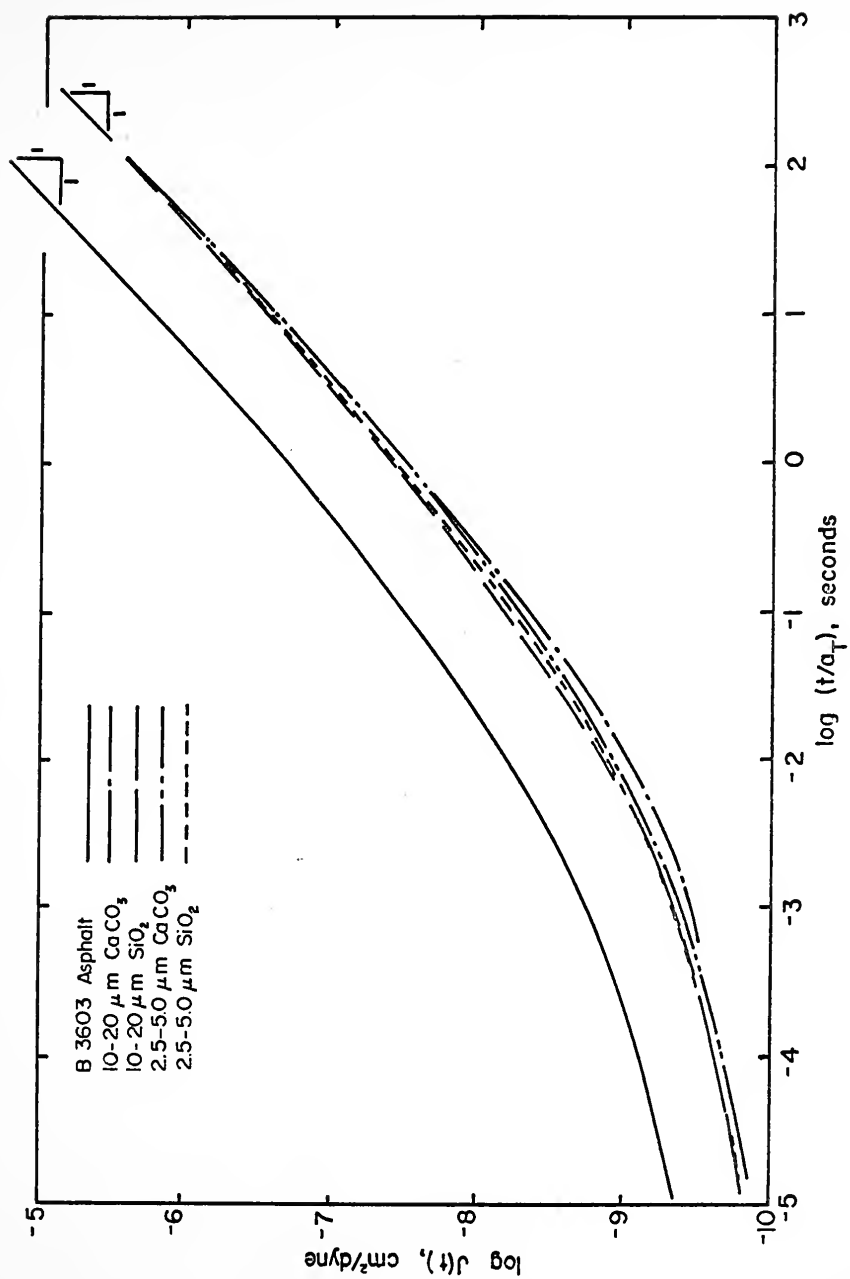


Figure 16. Creep Compliance, SiO_2 and CaCO_3 Mixtures, B-3603 Asphalt.

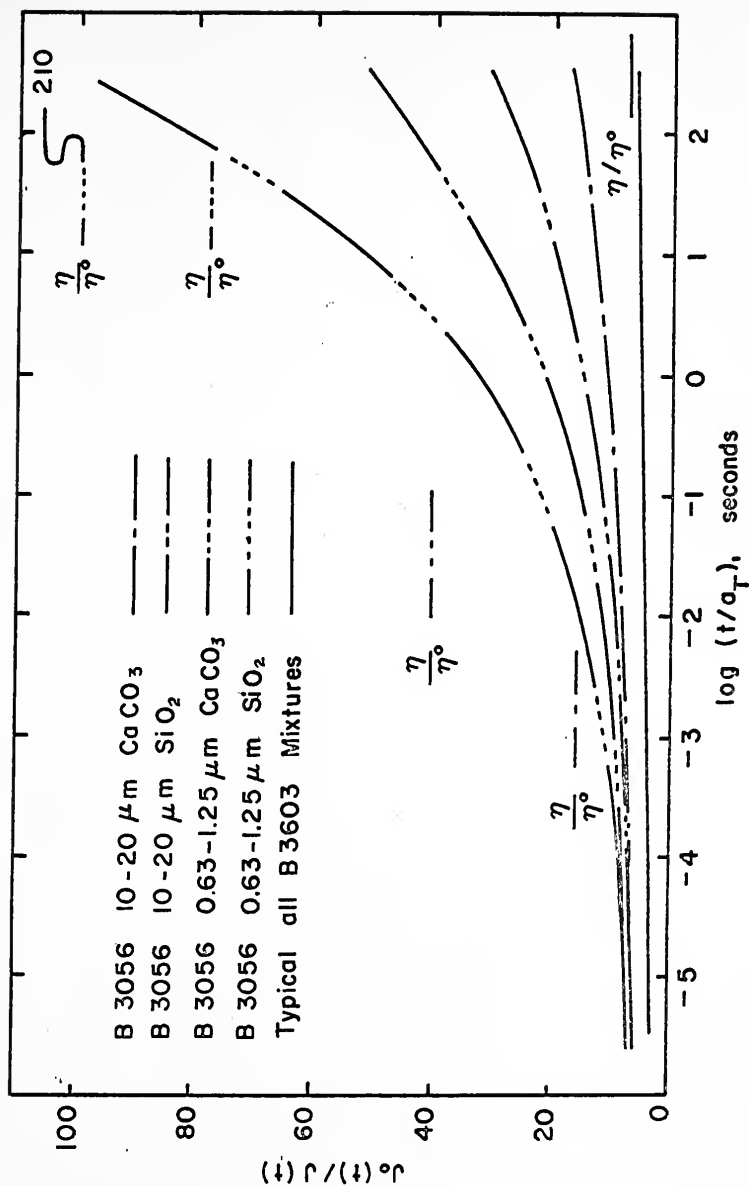


Figure 17. Ratio of Unfilled Filled Compliance, B-3056 and B-3603 Mixtures

